CLAIMS

1. A non-aqueous electrolytic solution comprising an electrolyte salt in a non-aqueous solvent for a lithium secondary battery, wherein the non-aqueous electrolytic solution further contains a vinylene carbonate compound represented by the formula (I) in an amount of 0.01 to 10 wt.%, and at least one alkyne compound represented by the formula (II), (III), (IV), (V), (VI), or (VII) in an amount of 0.01 to 10 wt.%:

10

in which each of R^1 and R^2 independently is a hydrogen atom or an alkyl group having 1 to 4 carbon atoms;

$$R^{3}-C \equiv C - \left(C - \frac{1}{C} \right)_{x} OY^{1}$$
(11)

in which each of R³ to R⁵ independently is a hydrogen atom, an alkyl group having 1 to 12 carbon atoms, a cycloalkyl group having 3 to 6 carbon atoms, or an aryl group having 6 to 12 carbon atoms, or R⁴ and R⁵ are combined with each other to form a cycloalkylene group having 3 to 6 carbon atoms; x is 1 or 2; and Y¹ is -COOR²⁰, -COR²⁰, or -SO₂R²⁰, wherein R²⁰ is a hydrogen atom, an alkyl group having 1 to 12 carbon atoms, a

cycloalkyl group having 3 to 6 carbon atoms, or an aryl group having 6 to 12 carbon atoms;

$$Y^{2}O \xrightarrow{R^{6}} C = C \xrightarrow{R^{8}} C \xrightarrow{R^{9}} OY^{3}$$

$$(111)$$

in which each of R⁶ to R⁹ independently is a hydrogen atom, an alkyl group having 1 to 12 carbon atoms, a cycloalkyl group having 3 to 6 carbon atoms, or an aryl group having 6 to 12 carbon atoms, or R⁶ and R⁷ or R⁸ and R⁹ are combined with each other to form a cycloalkylene group having 3 to 6 carbon atoms; x is 1 or 2; Y^2 is $-COOR^{21}$, $-COR^{21}$, or $-SO_2R^{21}$, wherein R^{21} is a hydrogen 10 atom, an alkyl group having 1 to 12 carbon atoms, a cycloalkyl group having 3 to 6 carbon atoms, or an aryl group having 6 to 12 carbon atoms; and Y³ is $-COOR^{22}$, $-COR^{22}$, or $-SO_2R^{22}$, wherein R^{22} is a hydrogen atom, an alkyl group having 1 to 12 carbon atoms, a 15 cycloalkyl group having 3 to 6 carbon atoms, or an aryl group having 6 to 12 carbon atoms;

in which each of R^{10} to R^{13} independently is a hydrogen atom, an alkyl group having 1 to 12 carbon atoms, a cycloalkyl group having 3 to 6 carbon atoms, or an aryl group having 6 to 12 carbon atoms, or R^{10} and R^{11} or R^{12}

and R^{13} are combined with each other to form a cycloal-kylene group having 3 to 6 carbon atoms; x is 1 or 2; Y^4 is $-COOR^{23}$, $-COR^{23}$, or $-SO_2R^{23}$, wherein R^{23} is a hydrogen atom, an alkyl group having 1 to 12 carbon atoms, a cycloalkyl group having 3 to 6 carbon atoms, or an aryl group having 6 to 12 carbon atoms; and Y^5 is $-COOR^{24}$, $-COR^{24}$, or $-SO_2R^{24}$, wherein R^{24} is a hydrogen atom, an alkyl group having 1 to 12 carbon atoms, a cycloalkyl group having 3 to 6 carbon atoms, or an aryl group having 6 to 12 carbon atoms;

5

10

15

$$R^{14}$$
— $C = C - (C - x) = C - R^{15} = C - R^{19} = C = C - R^{19} = C$

in which each of R^{14} to R^{19} independently is a hydrogen atom, an alkyl group having 1 to 12 carbon atoms, a cycloalkyl group having 3 to 6 carbon atoms, or an aryl group having 6 to 12 carbon atoms, or R^{15} and R^{16} or R^{17} and R^{18} are combined with each other to form a cycloalkylene group having 3 to 6 carbon atoms; and x is 1 or 2;

in which each of R²⁵ to R²⁷ independently is a hydrogen 20 atom, an alkyl group having 1 to 12 carbon atoms, a cycloalkyl group having 3 to 6 carbon atoms, an aryl group having 6 to 12 carbon atoms, or an aralkyl group having 7 to 12 carbon atoms, or R²⁶ and R²⁷ are combined with each other to form a cycloalkylene group having 3 to 6 carbon atoms; x is 1 or 2; W is sulfinyl, sulfonyl, or oxalyl; and Y⁶ is an alkyl group having 1 to 12 carbon atoms, an alkenyl group having 2 to 12 carbon atoms, an alkynyl group having 2 to 12 carbon atoms, a cycloalkyl group having 3 to 6 carbon atoms, an aryl group having 6 to 12 carbon atoms, or an aralkyl group having 7 to 12 carbon atoms;

$$R^{28} - \left(\begin{array}{c} \\ \end{array} \right)_{p} R^{29}$$
 (VII)

10

15

5

in which R^{28} is an alkyl group having 1 to 12 carbon atoms, a cycloalkyl group having 3 to 6 carbon atoms, or an aryl group having 6 to 12 carbon atoms; R^{29} is a hydrogen atom, an alkyl group having 1 to 12 carbon atoms, a cycloalkyl group having 3 to 6 carbon atoms, or an aryl group having 6 to 12 carbon atoms; and p is 1 or 2.

- The non-aqueous electrolytic solution of claim
 wherein the non-aqueous electrolytic solution contains
 the vinylene carbonate compound in an amount of 0.05 to 5 wt.%.
- The non-aqueous electrolytic solution of claim
 wherein the non-aqueous electrolytic solution contains
 the vinylene carbonate compound in an amount of 0.1 to 3 wt.%.

- 4. The non-aqueous electrolytic solution of claim 1, wherein the non-aqueous electrolytic solution contains the alkyne compound in an amount of 0.05 to 5 wt.%.
- 5. The non-aqueous electrolytic solution of claim 1, wherein the non-aqueous electrolytic solution contains the alkyne compound in an amount of 0.1 to 3 wt.%.
- 6. The non-aqueous electrolytic solution of claim
 10 1, wherein the vinylene carbonate compound is vinylene
 carbonate.
- 7. The non-aqueous electrolytic solution of claim
 1, wherein the alkyne compound is 2-propynyl methyl car15 bonate, 2-propynyl methanesulfonate, 2-butynylene
 bis(methyl carbonate), 2-butynylene
 bis(methanesulfonate), 2,4-hexadiynylene bis(methyl carbonate), di(2-propynyl) carbonate, di(2-propynyl) sulfite, di(2-propynyl) oxalate, phenylacetylene, ethyl 2propynyl oxalate, 2-propynyl formate, 2-butynylene diformate or 2,4-hexadiynylene diformate.

The non-aqueous electrolytic solution of claim 1, wherein the non-aqueous electrolytic solution further contains an aromatic compound in an amount of 0.1 to 5 wt.%, said aromatic compound being selected from the 5 group consisting of cyclohexylbenzene, a fluorocyclohexylbenzene compound, biphenyl, terphenyl, diphenyl ether, 2-fluorophenyl phenyl ether, 4-fluorophenyl phenyl ether, fluorobenzene, difluorobenzene, 2-fluorobiphenyl, 4-fluorobiphenyl, 2,4-difluoroanisole, tert-butylbenzene, 1,3-di-tert-butylbenzene, 1-fluoro-4-tert-butylbenzene, tert-pentylbenzene, tert-butyl biphenyl, tert-pentyl biphenyl, a partially hydrogenated o-terphenyl, a partially hydrogenated m-terphenyl and a partially hydrogenated pterphenyl.

15

10

The non-aqueous electrolytic solution of claim 9. 1, wherein the non-aqueous electrolytic solution further contains a mixture having a weight ratio of 50:50 to 10:90 in a total amount of 0.1 to 5 wt.%, said mixture being selected from the group consisting of a mixture of 20 biphenyl and cyclohexylbenzene, a mixture of cyclohexylbenzene and tert-butylbenzene, a mixture of cyclohexylbenzene and tert-pentylbenzene, a mixture of biphenyl and fluorobenzene, a mixture of cyclohexylbenzene and fluorobenzene, a mixture of 2,4-difluoroanisole and cyclohexyl-25 benzene, a mixture of cyclohexylbenzene and 1-fluoro-4tert-butylbenzene, a mixture of cyclohexylbenzene and a fluorocyclohexylbenzene compound, a mixture of a fluorocyclohexylbenzene compound and fluorobenzene, and a mix-30 ture of 2,4-difluoroanisole and a fluorocyclohexylbenzene compound.

- 10. A lithium secondary battery comprising a positive electrode, a negative electrode and a non-aqueous electrolytic solution, wherein the positive electrode comprises lithium mixed oxide, wherein the negative electrode comprises a material capable of absorbing and releasing lithium, and wherein the non-aqueous electrolytic solution is the solution defined in claim 1.
- 11. A lithium secondary battery comprising a positive electrode, a negative electrode and a non-aqueous electrolytic solution, wherein the positive electrode is a positive electrode composition layer having a density in the range of 3.2 to 4.0 g/cm³ provided on aluminum foil, said positive electrode layer composition layer comprising lithium mixed oxide, and wherein the non-aqueous electrolytic solution is the solution defined in claim 1.
- 12. A lithium secondary battery comprising a positive electrode, a negative electrode and a non-aqueous
 electrolytic solution, wherein the negative electrode
 comprises a negative electrode composition layer having a
 density in the range of 1.3 to 2.0 g/cm³ provided on copper foil, said negative electrode layer composition layer
 comprising a material capable of absorbing and releasing
 lithium, and wherein the non-aqueous electrolytic solution is the solution defined in claim 1.

13. A lithium secondary battery comprising a positive electrode, a negative electrode and a non-aqueous electrolytic solution, wherein the positive electrode comprises a positive electrode composition layer having a density in the range of 3.2 to 4.0 g/cm³ provided on aluminum foil, said positive electrode layer composition layer comprising lithium mixed oxide, wherein the negative electrode comprises a negative electrode composition layer having a density in the range of 1.3 to 2.0 g/cm³ provided on copper foil, said negative electrode layer composition layer comprising a material capable of absorbing and releasing lithium, and wherein the non-aqueous electrolytic solution is the solution defined in claim 1.

10